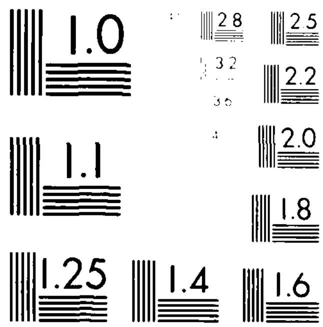
AD-A156 135 NOISE IN SILVER BETA ALUMINA CERAMICS(U) UTAH UNIV SALT LAKE CITY DEPT OF PHYSICS S W SMITH ET AL. SEP 85 TR-6 N00014-82-K-0603 1/1

UNCLASSIFIED F/G 9/1 NL


1.0
2.8
2.5
3.2
2.2
2.0
1.1
1.8
1.25
1.4
1.6

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER Six (6)	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER 5-28242
4. TITLE (and Subtitle) Noise in Silver β" Alumina Ceramics		5. TYPE OF REPORT & PERIOD COVERED Technical Report #6
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Steven W. Smith and James J. Brophy		8. CONTRACT OR GRANT NUMBER(s) N00014-82-K-0603
9. PERFORMING ORGANIZATION NAME AND ADDRESS Physics Department, University of Utah Salt Lake City, Utah 84112		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
11. CONTROLLING OFFICE NAME AND ADDRESS Leader, Chemistry Div., Associate Director of Mathematics & Physical Sciences, Office of Naval Research, 800 N. Quincy St., Arlington, VA 22217		12. REPORT DATE September 1985
		13. NUMBER OF PAGES Nine (9)
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) Office of Naval Research Resident Representative University of California 239 Campbell Hall Berkeley, CA 94720		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release or sale.
Distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

-

18. SUPPLEMENTARY NOTES

-

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Diffusion noise, conductivity fluctuations, superionic conductors, beta alumina.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Voltage fluctuations at ohmic electrodes to silver β" alumina ceramic samples are observed both at contacts and in the bulk over the frequency interval 10^{-3} to 10^{4} Hz. Contact noise power in the absence of current varies as f^{-2} at low frequencies and is dominated by Nyquist noise of the sample at frequencies greater than 100 Hz. Bulk current noise measured at transverse contacts has a $f^{-3/2}$ power spectrum and increases with the square of the current. Low-frequency contact noise, sample Nyquist noise and bulk current noise are all thermally activated with activation energies of -0.96, 0.14 and 0.61 eV, respectively. (over)

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE
S/N 0102-LF-014-6601

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

20. Abstract (continued)

These experimental results are very similar to those previously reported for sodium β" alumina ceramics.

Office of Naval Research
Attn: Code 413
800 N. Quincy Street
Arlington, Virginia 22217

Dr. Bernard Douda
Naval Weapons Support Center
Code 5042
Crane, Indiana 47522

Dr. David Young
Code 334
NORDA
NSTL, Mississippi 39529

Commander, Naval Air Systems
Command
Attn: Code 310C (H. Rosenwasser)
Washington, D.C. 20360

Naval Weapons Center
Attn: Dr. A. B. Amster
Chemistry Division
China Lake, California 93555

Naval Civil Engineering Lab
Attn: Dr. R. W. Drisko
Port Hueneme, California 93401

Scientific Advisor
Commandant of the Marine Corps
Code RD-1
Washington, D.C. 20380

Defense Technical Information Ce
Building 5, Cameron Station
Alexandria, Virginia 22314

U.S. Army Research Office
Attn: CRD-AA-IP
P.O. Box 12211
Research Triangle Park, NC 27709

DTNSRDC
Attn: Dr. G. Bosmajian
Applied Chemistry Division
Annapolis, Maryland 21401

Mr. John Boyle
Materials Branch
Naval Ship Engineering Center
Philadelphia, Pennsylvania 19112

Dr. William Tolles
Superintendent
Chemistry Division, Code 6100
Naval Research Laboratory
Washington, D.C. 20375

Naval Ocean Systems Center
Attn: Dr. S. Yamamoto
Marine Sciences Division
San Diego, California 91232

OFFICE OF NAVAL RESEARCH

Cntract #N00014-82-K-0603

TECHNICAL REPORT No. 6

NOISE IN SILVER β'' ALUMINA CERAMICS

by

Steven W. Smith and James J. Brophy

Prepared for Presentation

at the

8th International Conference on Noise in Physical Systems
Rome, Italy

Department of Physics
University of Utah
Salt Lake City, Utah 84112

September 1985

Office of Naval Research
Attn: Code 413
800 N. Quincy Street
Arlington, Virginia 22217

Dr. Bernard Douda
Naval Weapons Support Center
Code 5042
Crane, Indiana 47522

Dr. David Young
Code 334
NORDA
NSTL, Mississippi 39529

Naval Weapons Center
Attn: Dr. A. B. Amster
Chemistry Division
China Lake, California 93555

Commander, Naval Air Systems Command
Attn: Code 310C (H. Rosenwasser)
Washington, D.C. 20360

Naval Civil Engineering Lab
Attn: Dr. R. W. Drisko
Port Hueneme, California 93401

Scientific Advisor
Commandant of the Marine Corps
Code RD-1
Washington, D.C. 20380

U.S. Army Research Office
Attn: CRD-AA-IP
P.O. Box 12211
Research Triangle Park, NC 27709

Defense Technical Information Ce
Building 5, Cameron Station
Alexandria, Virginia 22314

DTNSRDC
Attn: Dr. G. Bosmajian
Applied Chemistry Division
Annapolis, Maryland 21401

Mr. John Boyle
Materials Branch
Naval Ship Engineering Center
Philadelphia, Pennsylvania 19112

Dr. William Tolles
Superintendent
Chemistry Division, Code 6100
Naval Research Laboratory
Washington, D.C. 20375

Naval Ocean Systems Center
Attn: Dr. S. Yamamoto
Marine Sciences Division
San Diego, California 91232

NOISE IN SILVER β'' ALUMINA CERAMICS

Steven W. Smith and James J. Brophy

University of Utah
Salt Lake City, Utah 84112

Voltage fluctuations at ohmic electrodes to silver β'' alumina ceramic samples are observed both at contacts and in the bulk over the frequency interval 10^{-3} to 10^{4} Hz. Contact noise power in the absence of current varies as f^{-2} at low frequencies and is dominated by Nyquist noise of the sample at frequencies greater than 100 Hz. Bulk current noise measured at transverse contacts has a $f^{-3/2}$ power spectrum and increases with the square of the current. Low-frequency contact noise, sample Nyquist noise and bulk current noise are all thermally activated with activation energies of -0.96, 0.14 and 0.61 eV, respectively. These experimental results are very similar to those previously reported for sodium β'' alumina ceramics.

1. INTRODUCTION

Electrical noise voltages attributed to diffusion noise of the mobile ions have been observed in superionic sodium β'' alumina ceramics[1]. This interpretation leads to conclusions that only a small fraction of the mobile ions participate in the noise process and that the granularity does not influence the diffusion noise magnitude. It is of interest to examine conductivity fluctuations in silver β'' alumina ceramics to determine the influence of the nature of the mobile ions on the noise processes. This study is facilitated by the ease with which the mobile ions can be exchanged in the β'' alumina structure[2].

2. EXPERIMENTAL PROCEDURE

Commercial sodium β'' alumina ceramic (90.4% Al_2O_3, 8.85% Na_2O, 0.75% Li_2O) specimens[3], 1x1x0.3 cm^3, are converted to silver β'' alumina by ion exchange in molten 50% $AgNO_3/NaNO_3$ at 300° C for 8 hours. Weight change of the samples indicates 98% of the mobile sodium ions are replaced by silver ions. Ohmic contacts are 5-M $AgNO_3$ solution in water or 0.05-M glycerin. Silver amalgam contacts are ohmic, but have very high resistance[4]. The corners of the sample are sealed into the sides of four plastic test tubes containing the liquid electrode material to provide diagonally opposing corner current terminals and transverse noise contacts. This is essentially the same configuration used to study sodium β'' alumina ceramics[1].

The transverse noise contacts are connected to the input of a PAR 113 preamplifier. Digital analysis of the preamplifier output is accomplished using A/D conversion and a FFT program developed for the Apple IIe personal computer[5]. The system operates over the frequency interval 10^{-4} to 10^4 Hz and is calibrated using the Nyquist noise of resistances ranging from 10^4 to $2x10^8$ ohm. Sample current terminals are connected to a filtered battery source through a 10^5 ohm noiseless series resistor.

3. NOISE RESULTS

Typical noise spectra, Figure 1, show con-

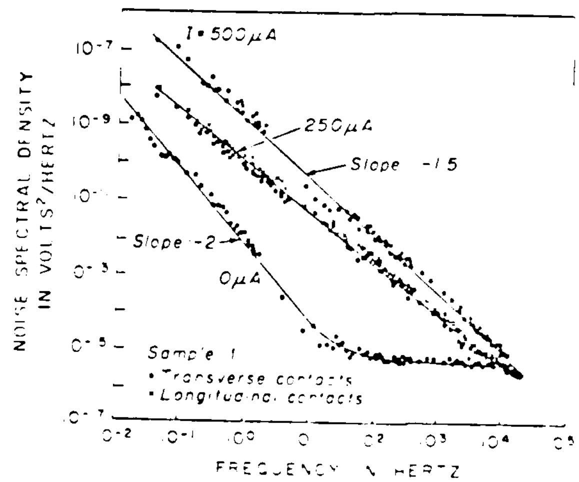


Figure 1. Noise spectra of silver β" alumina ceramic.

tact noise[6] in the absence of current and current noise characterized by an $f^{-1.5}$ trend. The slopes of the spectra tend to be smaller, $f^{-1.3}$, at low currents. At high frequencies the observed noise is accounted for by Nyquist noise of the sample resistance calculated from the linear current-voltage characteristic. In general, all three noise properties are very similar to sodium β" alumina[1]. The

range of current noise magnitudes exhibited by the silver samples overlap those of the sodium conductors while the Nyquist noise level is greater, in agreement with the greater resistivity of silver β" alumina[2]. Similarly, contact noise levels tend to be greater, although still sufficiently low to permit reliable current noise measurements.

The contact noise levels of aqueous and glycerin solutions are identical after each ages for several hours. Both contact materials exhibit noise levels in excess of the 2×10^{8} ohm preamplifier input resistance Nyquist noise at frequencies below 10^{-1} Hz and the spectral shapes are near f^{-2}. These characteristics are attributed to non-equilibrium chemical reaction noise at the sample-electrode interface[6]. Since both electrode solutions have the same noise level, chemical contact noise must be associated with the mobile silver ions.

Current noise in silver β" alumina is much more stable with respect to time and current than is the case for sodium β" specimens. Relatively minor changes attributable to electrochemical effects[1] are observed. Also, transverse and longitudinal (two-terminal) noise levels are the same for low-noise contacts. This means that contact current noise levels are small compared to bulk conductivity fluctuations.

Both the stability and the absence of contact current noise suggest that silver β" alumina is a better material than sodium β" alumina to use in the study of noise in superionic conductors.

A few samples inadvertently heated to high temperatures (800°C) for several hours experienced an increase in resistivity of four to five times, together with a darkening of the surface. This behavior has been previously reported[7], but no chemical or structural change has been detected to account for the increase in resistivity. Noise properties of such "darkened" samples are not noticeably different from normal specimens, although the temperature dependence changes somewhat.

As in the case of sodium β" alumina, the various noise processes are thermally activated, Figure 2. The activation energy for Nyquist noise of the normal sample is a little lower than literature values[2,7], but the increase upon darkening is consistent with conductivity data[7]. The current noise activation energy is about one-half of that for sodium β" alumina, while the activation energy for contact noise is significantly greater. This is consistent with the greater contact noise level of silver β" alumina and a thermally-activated chemical reaction at the contact.

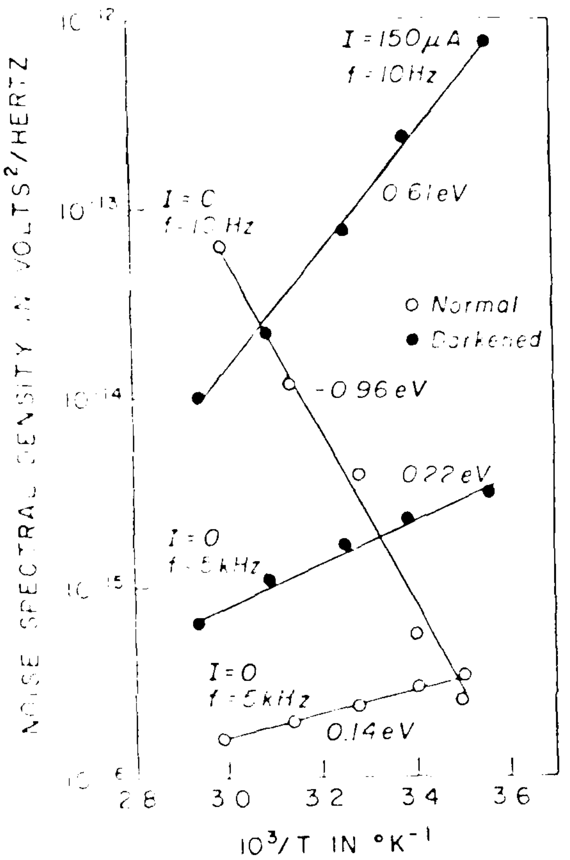


Figure 2. Temperature dependence of Nyquist noise (0 μA, 5 kHz) current noise (150 μA, 10 Hz), and contact noise (0 μA, 10 Hz) of normal and darkened silver β" alumina ceramic.

4. DISCUSSION

As in the case of sodium β" alumina ceramics, the $f^{-1.5}$ spectral shape suggests

diffusion noise, for which the noise voltage spectral density can be written as[8]

$$\frac{S(V,f)}{V^2} = 4\,\frac{\langle \Delta N^2 \rangle}{N}\left(\frac{D}{2L^2}\right)^{1/2} \omega^{-3/2} \qquad (1)$$

where V is the dc voltage across the sample, $\langle \Delta N^2 \rangle$ and N are the variance and average number of diffusing ions, D is the diffusion constant, L is a characteristic length, and ω is the angular frequency. This expression is valid above a characteristic frequency ω_0,

$$\omega_0 = 2D/L^2 \qquad (2)$$

Below ω_0 the spectrum flattens, becoming constant in the case of diffusion in three dimensions.

Taking $D=10^{-7}$ cm^2/sec at room temperature[2,7], the turnover frequency calculated from Equation 2 is equal to $3x10^{-8}$ Hz if L is the sample length and $1.3x10^{-1}$ Hz if L is equal to the average gain size[9], $5x10^{-4}$ cm. No departure from the $f^{-1.5}$ trend is noticed down to 10^{-3} Hz, so that if appears that the granular structure does not influence the diffusion noise. This is consistent with data from sodium β'' alumina ceramics[1].

As in the case of sodium β" alumina[1], observed noise levels are very much larger than those predicted by Equation 1. Furthermore, the activation energy seen in Figure 2 is greater than can be accounted for by Equation 1, unless the number of diffusing ions is thermally activated. This approach leads to satisfactory numerical agreement in the case of sodium β" alumina[1], but does not appear to apply to silver β" alumina.

5. CONCLUSIONS

These experimental results show that conductivity fluctuations in silver β" alumina ceramics are very similar to those previously observed in sodium β" alumina. The spectral shape suggests diffusion noise and the noise does not appear to be influenced by the granularity of the samples. It is not possible to account quantitatively for the observed magnitude of the noise by assuming that the active ion density is thermally activated, as in the case of sodium β" alumina.

6. ACKNOWLEDGMENTS

The authors express their appreciation to G. R. Miller and J. M. Viner for many helpful suggestions and advice. This work is supported in part by the Office of Naval Research.

7. REFERENCES

1) James J. Brophy and Steven W. Smith, J. Appl. Phys., 58, 700 (1958).

2) J. L. Briant and G. C. Farrington, J. Solid State Chem., 33, 385 (1980).

3) Obtained from Ceramatec, Inc., Salt Lake City, Utah 84115.

4) James J. Brophy, "Noise in Physical Systems and 1/f Noise", M. Savelli, G. Lecoy, and J-P. Nougier (eds), Elsevier Science Publishers, B.V., Amsterdam, (1983) p. 351.

5) Steven W. Smith, Rev. Sci. Inst., 56, 159 (1985).

6) James J. Brophy and Steven W. Smith, J. Appl. Phys. 56, 801 (1984).

7) M. W. Breiter and B. Dunn, Sol. State Ionics, 9&10, 227 (1983).

8) K. M. Van Vliet and J. R. Fassett, "Fluctuation Phenomena in Solids", R. E. Burgess (ed), Academic Press, New York (1965), p. 268.

9) A. D. Jatkar, I. B. Cutler, A. V. Virkar, and R. S. Gordon, "Processing of Crystalline Ceramics", H. Palmour, R. F. Davis and T. M. Hare (eds), Plenum Publishing, New York (1978), p. 421.

END

FILMED

8-85

DTIC